Title	Lamellar and Crystalline-Core Thicknesses of Poly(3-oxotrimethylene)(POK) Crystallized Epitaxially on Alkali Halides / Multi-Axial Deformations of End-linked Polydimethylsiloxane Model Networks (STATES AND STRUCTURES - Polymer Condensed States)
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States and Structures - Polymer Condensed States -

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Scope of Research

Attempts have been made to elucidate the molecular arrangement and the mechanism of structural formation/change in crystalline polymer solids, polymer gels and elastomers, polymer liquid crystals, and polymer composites, mainly by electron microscopy and/or X-ray diffraction/scattering. The major subjects are: synthesis and structural analysis of polymer composite materials, preparation and characterization of polymer gels and elastomeric materials, structural analysis of crystalline polymer solids by direct observation at molecular level resolution, and *in situ* studies on structural formation/change in crystalline polymer solids.

Research Activities (Year 2001)

Presentations

Morphological investigation on the formation of fibrillar structure for PEN fibers, Yoshioka T, Tsuji M, Kohjiya S, et al., Annual Meeting, Soc. of Fib. Sci. and Technol., Jpn., 6-8 June, and related 1 presentation.

Structure analysis of polymer solids mainly by transmission electron microscopy, Tsuji M, Symposium on Macromolecules, Soc. Polym. Sci., Jpn., 12-14 September.

Biaxial deformation of endlinked model polydimethylsiloxane networks, Kawamura T, Urayama K, Kohjiya S, Annual Meeting, Soc. Polym. Sci., Jpn., 24 May, and related 5 presentations.

Swelling behavior of liquid crystalline gels in liquid crystalline solvents, Okuno Y, Kawamura T, Urayama K, Kohjiya S, Symposium on Macromolecules, Soc. Polym. Sci., Jpn., 13 September.

Electromechanical properties of thin polymer layers, Urayama K, Neher D (Potsdam Univ.), et al., Symposium on Macromolecules, Soc. Polym. Sci., Jpn., 13 September, and related 3 presentations.

Grants

Kohjiya S, Urayama K, Murakami S, Ikeda Y, Role of polymers in all solid-state ionic devices, Grant-in-Aid for Scientific Research, Priority Area (B), 1 April 1999 - 31 March 2004

Kohjiya S, Tsuji M, Urayama K, Direct observation of amorphous polymer network structures by TEM, Grant-in-Aid for Scientific Research, (B)(2), 1 April 2001 - 31 March 2003

Urayama K, Dynamics of guest polymers in host polymer networks, Grant-in-Aid for Scientific Research, Encouragement of Young Scientists, 1 April 2001 - 31 March 2003

Topics

Lamellar and Crystalline-Core Thicknesses of Poly(3-oxotrimethylene) (POK) Crystallized Epitaxially on Alkali Halides

Edge-on lamellar crystals of POK were isothermally grown from a dilute solution in nitrobenzene epitaxially onto KI at various temperatures (T_c = 150–190 ºC). The resulting crystals were observed by transmission electron microscopy (TEM) in bright-field (BF) and dark-field (DF) modes and by high-resolution (HR) TEM. The DF images obtained by conventional TEM and the (110) lattice images obtained by HR-TEM showed that the crystalline-core thickness of each lamella is inevitably smaller than the corresponding lamellar thickness. Figure 1 shows the T_c-dependence of the crystalline-core thickness obtained by DF- (circle) and HR-TEM (triangle), and also that of the lamellar thickness obtained by BF-TEM of the edge-on lamellae (square) and by small-angle X-ray scattering of the "single crystal" mats (+). Both of the thicknesses increase with increasing T_c. For the crystals grown at any T_c, the crystalline-core thickness is 50–60 % of the corresponding lamellar thickness. Thus the POK lamella has a surface layer (20–25 % of the lamellar thickness) containing folds on each basal side of the lamella.

1. M. Fujita, et al., *Macromolecules*, **34**, 6147 (2001)
2. M. Fujita, et al., *Macromolecules*, **34**, 7724 (2001)

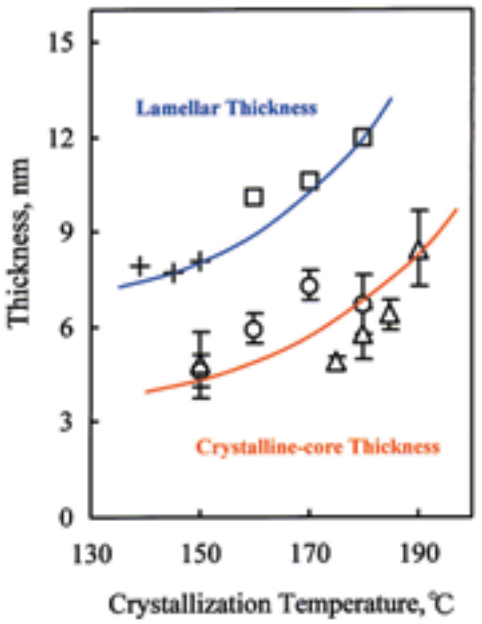

Figure 1.

Multi-Axial Deformations of End-linked Polydimethylsiloxane Model Networks

Molecular interpretation of rubber elasticity of cross-linked amourphous polymer networks still remains incomplete due to the complicated network topology, especially, entanglement couplings of different network chains. A large number of theoretical models with different treatments of entanglement effects have been proposed. We have prepared a model polydimethylsiloxane network which has a well-characterized network chain length and junction functionality via end-linking method. Using this model network, we have carried out the biaxial deformation experiment which achieves all accessible pure homogeneous strains [1,2]. The data obtained enable us to identify the models which account for the entanglement effects most successfully and correctly [2]. Among the five molecular theories tested, the Edwards-Vilgis slip-link model [3] shows the most successful reproducibility over large portions of the experimental data, as shown in Figure 2.

1. T. Kawamura, et al., *Macromolecules*, **34**, 8252 (2001)
2. K. Urayama, et al., *Macromolecules*, **34**, 8261 (2001)
3. S. F. Edwards, et al., *Rep. Prog. Phys.*, **51**, 243 (1988)

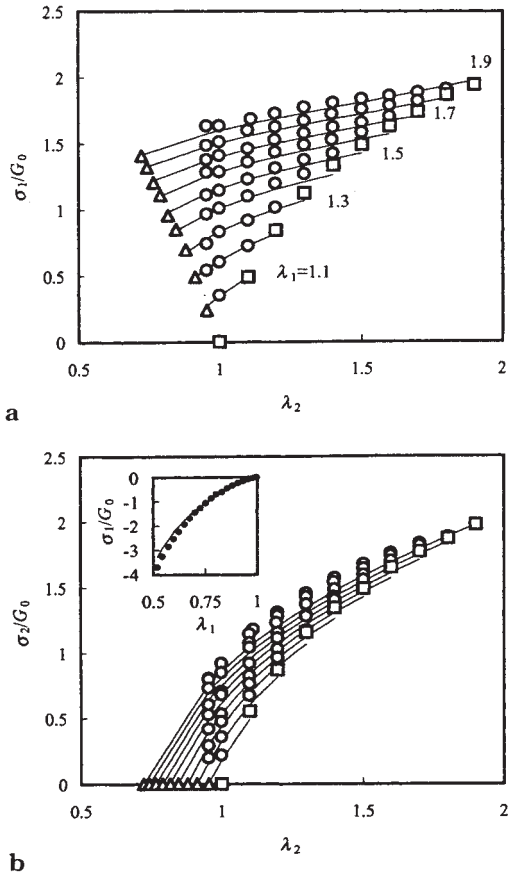

Figure 2. Comparisons of the experimental reduced principal stresses (a) σ_1/G_o and (b) σ_2/G_o - principal elongations (λ_1 and λ_2) relations with the predictions of the slip-link model (solid lines). The inset of part b shows the comparisons for uniaxial compression.